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3

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RECENT ADVANCES IN SEGMENTED GAMMA SCANNER ANALYSIS

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ABSTRACT

The segmented gamma scanner (SGS) is used in many facilities to assay low-density scrap and waste generated in the facilities. The procedures for using the SGS can cause a negative bias if the sample does not satisfy the assumptions made in the method. Some process samples do not comply with the assumptions. This paper discusses the effect of the presence of lumps on the SGS assay results, describes a method to detect the presence of lumps, and describes an approach to correct for the lumps. Other recent advances in SGS analysis are also discussed.

I. INTRODUCTION

Segmented gamma scanning is a procedure that was developed by Los Alamos National Laboratory in the early 1970s.[1] The procedure was developed to assay the special nuclear material (SNM) contained in low-density scrap and waste, such as paper wipes, gloves, and plastic containers, by measuring the gamma emission rate. The instrument based on this procedure, the segmented gamma scanner (SGS), is widely used in the US DOE complex and in industry, and it is manufactured by commercial vendors. Recent developments have led to modifications in the hardware design[2] and in the analysis procedures. This paper will summarize the advances in SGS analysis procedures.

Quantitative gamma ray assays nominally require that the sample be homogeneous; however, most scrap and waste are highly heterogeneous. The traditional SGS procedures correct for minor heterogeneity in a low-density matrix, but assume the SNM is non self absorbing. However, most of the scrap and waste found in the facility do not satisfy the nonabsorbing assumption; with these samples, an assay with the SGS is usually biased.[1] The primary purpose of this paper is to discuss the bias caused by the samples containing lumps of SNM.

II. ASSAY PRINCIPLES

In the SGS procedure, three methods have traditionally been employed to reduce the effects of heterogeneity.[1] To improve the vertical homogeneity, the sample is assayed segment by segment. To improve the horizontal homogeneity, the sample is rotated during the assay of each segment. A transmission measurement is made on each segment to correct for the sample attenuation.

The segmentation along the vertical axis of the sample provides two benefits. It is an effective way of dealing with vertical nonuniformity in the assay samples, and it allows the use of a single assay geometry for all sample heights.

Rotating the sample is a way to reduce the adverse effect of radial inhomogeneities while maintaining the highest possible count rate. Achieving a high count rate requires the detector to be positioned as closely as possible to the sample, whereas a large ratio of sample diameter to detector distance introduces substantial count rate dependence upon radial inhomogeneities in the sample.

For each segment, the overall transmission is measured and then used to calculate an attenuation correction. The fundamental assumptions here are that the mixture of SNM and matrix is reasonably uniform and that the particles of SNM are small enough that self-attenuation within the individual gamma-ray emitting particles can be ignored. In summary, this assumption states that the sample attenuation is characterized by a single linear attenuation coefficient.

III. SOURCES OF BIAS

Two sources of bias have been identified in the SGS procedure described above: the end effect and the lump effect. Both will result in a negatively biased assay.[1]

The end effect arises because only part of the gamma-ray beam from the transmission source is intercepted by the sample material as the top or bottom of the sample material moves across the collimator. A schematic representation of the end effect is shown in Fig. 1. An erroneously high transmission leads to an attenuation correction factor that is too small. Consequently, the corrected count from such a segment is low, which causes a negative bias in the assay results. It is possible to minimize the end effect by requiring that the sample depth be at least 6 times, but preferably more than 10 times, the segment thickness. However, in some operating facilities it is not practical to require that all the waste cans be filled to at least a specified height.

The lump effect describes the situation when the samples contain SNM in the form of lumps rather than being finely distributed. For example, if the material emitting the gamma rays consists of lumps of uranium metal embedded in a low-density matrix, the transmission measurement would tend to reflect the attenuation of the low-density matrix since it would dominate the area of the sample seen by the transmission source. However, the SNM gamma rays would be attenuated mainly in the lumps of uranium metal from which they arise. The attenuation correction factor would then be low, yielding an assay value below the actual value.

IV. RECENT ADVANCES IN SGS ANALYSIS

A. Method of Detection and Correction of Lumps

One of the possible methods to detect the presence of lumps is to assay the sample at multiple energies. If we assay the sample with several gamma-ray peaks of different energies, the assay at the lower energy will be more suppressed by the presence of lumps than will the assay at higher energy. This occurs because the self-absorption is greater for lower-energy gamma rays. This is explained graphically in Fig. 2 for the case of plutonium-bearing waste.

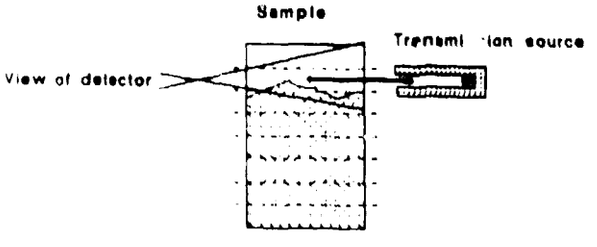

Fig. 1. This figure illustrates the cause of the end effect. The measured transmission of the end segment is usually too high.

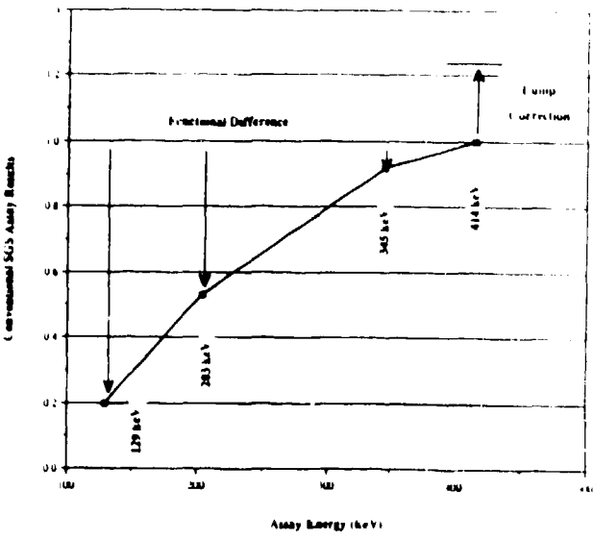

Fig. 2. This figure shows the conventional SGS assay of a plutonium sample containing lumps of SNM.

It is possible to assay the waste by means of several lower-energy peaks and the 414-keV gamma-ray peak of ^{239}Pu; the assays with the lower-energy peaks will be suppressed comparatively more than the assay with the 414-keV peak. The correction for lumps at the 414-keV energy is what is needed; what can be measured is the fractional differences between the peaks at 129 and 414 keV, between 203 and 414 keV, and between 345 and 414 keV. Each of these three fractional differences can be used to estimate a correction to the 414-keV result.

The effect of the lumps is also shown in Table I , which the traditional SGS assay of samples from the molten salt extraction (MSE) separation process is shown. These salts nominally contain chunks of plutonium. Notice that for the standard, the assay results from the various energies are in agreement. However, for the salt samples, the assay results in general increase with gamma ray energy, indicating the presence of lumps

To arrive at a lump correction, we assume that the lumps are in the form of spheres. The actual lumps may deviate from spherical shape, but if the lumps are randomly oriented in the sample, the spherical assumption may be quite appropriate.

The fraction of unscattered and unabsorbed gamma rays escaping from a sphere can be expressed by[4]

$$f = \frac{1}{2X}\left[1 - \frac{2}{X^2} + e^{-X}\left(\frac{2}{X} + \frac{2}{X^2}\right)\right],$$

TABLE I

TRADITIONAL SGS ASSAY OF MSE SALTS

Assay of ^{239}Pu based on the following gamma peak

Sample	129 keV	203 keV	345 keV	414 keV
ASH-1(standard)	19.48 ± 0.27	19.90 ± 0.42	19.72 ± 0.23	19.64 ± 0.09
RFMSE3	34.85 ± 0.24	43.60 ± 0.41	47.51 ± 0.26	48.19 ± 0.13
RFMSE5	54.96 ± 1.2	82.04 ± 0.93	109.19 ± 0.43	112.26 ± 0.24
ARF876700	150.9 ± 4.4	187.05 ± 2.6	198.7 ± 0.56	197.25 ± 0.31
XBLP-267	76.7 ± 0.7	104.5 ± 1.0	116.3 ± 0.5	116.7 ± 0.2
XBLP-120	78.5 ± 0.9	90.8 ± 0.9	91.9 ± 0.5	91.0 ± 0.2
XBLP-270	60.6 ± 0.6	81.6 ± 0.8	89.9 ± 0.4	89.8 ± 0.2
XBLP-300	108.1 ± 0.83	147.6 ± 0.9	163.98 ± 0.42	163.94 ± 0.21
XBLP-301	92.6 ± 0.9	134.86 ± 1.2	179.76 ± 0.57	186.0 ± 0.3

where X = μρD,
μ = mass attenuation coefficient,
ρ = density of sphere, and
D = diameter of sphere.

Using this expression, it is possible to calculate the fraction of gamma rays escaping from the sphere at different energies. The lump correction factor is defined as 1/F. Since this expression is nonlinear with respect to X, if the lumps span a range of sizes, the correction factor deduced from this expression may be slightly in error. The fractional differences and the corresponding lump correction factors are shown in Table II and in Fig. 3.

To determine the lump correction, the SGS system has to be calibrated to standards that are known to contain no lumps. With these calibration constants, the traditional SGS assay is performed at different energies. This assumes that the traditional corrections for matrix attenuation, branching ratios, and count rate effects have already been made. Based on the ratio of the results between a lower-energy assay and the 414-keV assay, a lump correction can be applied to the 414-keV assay. If the results agree within the measurement error, there will be no lump correction.

B. Extrapolation of Transmission

In the traditional SGS, for the ^{239}Pu assay, the transmission is measured at the 400-keV peak of ^{75}Se and assumed to be the same at the 414-keV peak of ^{239}Pu.

For ^{75}Se, there are other strong gamma rays at 279 keV and 136 keV; the transmissions

TABLE II

LUMP CORRECTION AT 414 keV

ρD (g/cm²)	414-129 Fract. Diff.	414-203 Fract. Diff.	414-345 Fract. Diff.	414 Lump Correction
0.5	0.4301	0.1584	0.0211	1.0518
1	0.6176	0.2769	0.0408	1.1053
1.5	0.7131	0.3671	0.0589	1.1502
2.0	0.7682	0.4365	0.0759	1.2165
2.5	0.8032	0.4909	0.0915	1.2744
3.0	0.8272	0.5319	0.1059	1.3115
3.5	0.8446	0.5686	0.1193	1.3319
4.0	0.8576	0.5968	0.1319	1.4556
5.0	0.8757	0.6393	0.1542	1.5828
6.0	0.8876	0.6692	0.1730	1.7150
7.0	0.8959	0.6909	0.1893	1.8512
8.0	0.9019	0.7072	0.2033	1.9916
9.0	0.9065	0.7199	0.2155	2.1358
10.0	0.9100	0.7294	0.2256	2.2836

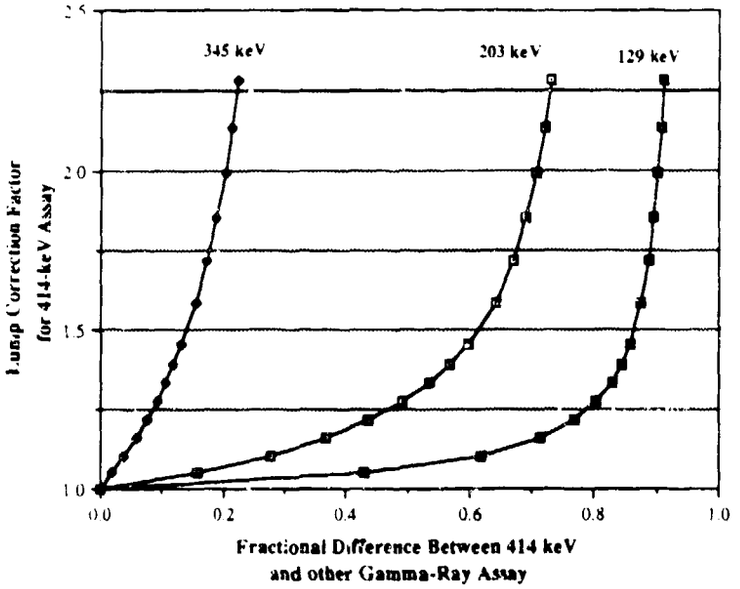

Fig. 3. The lump correction factor as a function of fractional differences between the peaks at 129 and 414 keV, 203 and 414 keV, and 345 and 414 keV.

of these gamma rays can be measured simultaneously with the 400-keV peak. With transmissions at three energies, it is possible to use a straight-line extrapolation, or to use a quadratic fit to calculate the transmission at 414 keV. In addition, to apply the lump correction for the plutonium assay, it is also necessary to calculate the transmission at 129 keV, 203 keV, and 345 keV.

The following is a comparison of the two methods of extrapolation and interpolation. A 1-cm cell of plutonium in a 3-M nitric acid solution of varying concentrations is assumed for the sample. Because the composition of the sample is known, it is possible to calculate the transmission for the 136-keV, 279-keV, and 400-keV peaks of ^{75}Se. It is also possible to calculate the transmission at the four plutonium peak energies: 129, 203, 345, and 414 keV. A straight-line fit is performed in the $\ell n[-\ell n(T)]$ vs $\ell n E$ space between 136 keV and 279 keV, and from this line the transmissions at 129 keV and 203 keV can be calculated. Similarly, a straight-line fit is also performed between 279 keV and 400 keV, and the transmissions at 345 keV and 414 keV are also calculated. The comparison between the correct transmission values for the plutonium energies and the interpolated values is summarized in Fig. 4. It can be noted that for low concentrations (<50 g/ℓ), both the straight line fit and the quadratic fit do quite well; at higher concentrations the quadratic fit is better in calculating the transmissions. Note that the extrapolation to 414 keV is accurate to ≤0.3% for both methods; either method will provide a better determination of the transmission at 414 keV than does the traditional SGS method.

C. Attenuation Correction Factor

In the traditional SGS procedures, the attenuation correction factor is based on the approximate formula

$$CF(T) = \frac{-K\ell n(T)}{1 - T^K} ,$$

where K is a constant = 0.823.[1,4] This correction factor is accurate to within 5% over a wide range of transmissions. While this correction factor is adequate when optimum accuracy is not required, it is possible to improve the accuracy by fitting a function of the form[4]

$$CF(T) = A + B\log(T) + C\,[\log(T)]^2 .$$

In a typical SGS geometry, A, B, and C can be determined to give CF(T) within 0.3% for $0.208 \le T \le 0.30$.

V. COMPARISON WITH DESTRUCTIVE ANALYSIS

Table I shows results for a traditional SGS assay of molten salts generated from the pyrochemical process. By assuming the lumps are uniform plutonium spheres, we have derived the method for the lump correction described above. This assumes that the assays at different gamma ray energies are independent and that

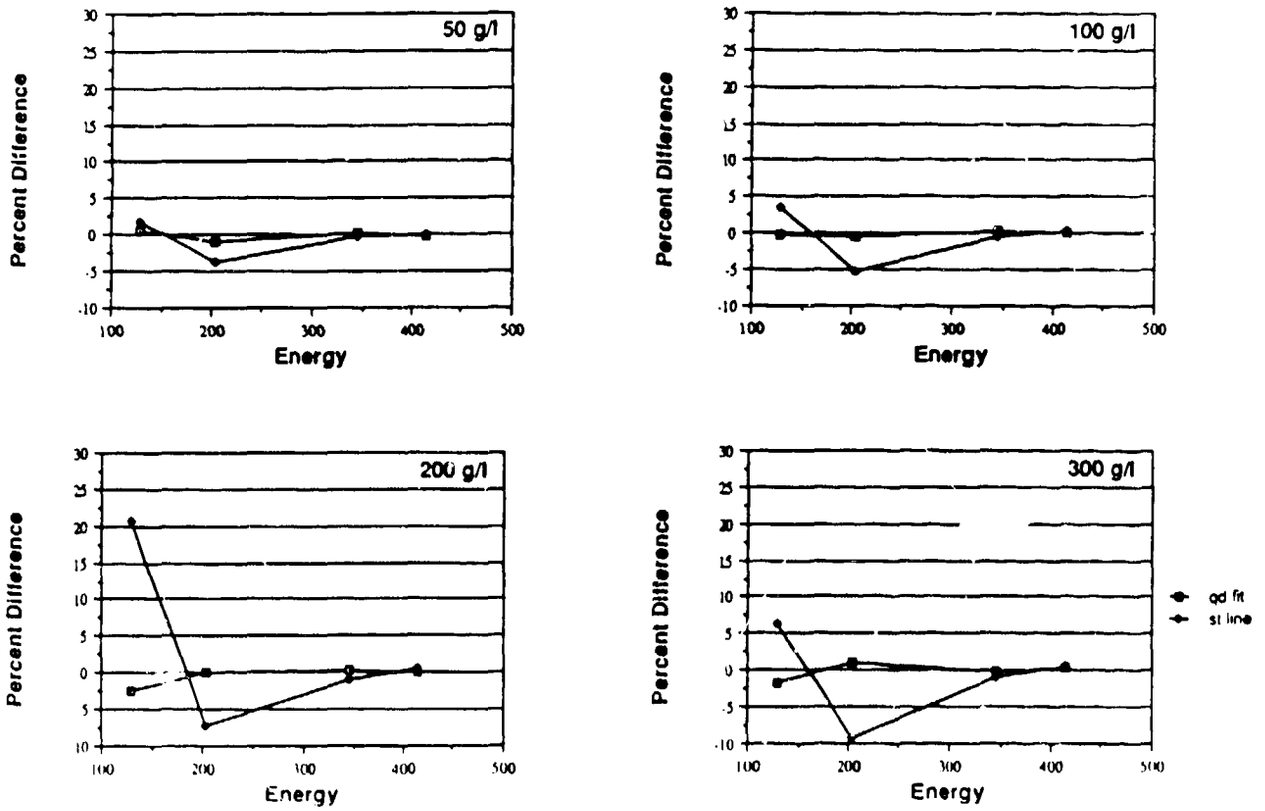

Fig. 4. Percent difference between the actual transmission and the interpolated value using a straight-line interpolation, and the percent difference using a quadratic fit.

TABLE III

ADVANCED SGS ASSAY OF MSE SALT

(Lump-corrected assays at 414 keV. Tabulated values are in g ^{239}Pu. The uncertainties are 1σ values estimated from counting statistics.)

Sample	414-keV Result Corrected by 129-keV Result	414-keV Result Corrected by 203-keV Result	414-keV Result Corrected by 345-keV Result	Lump Correction to 414-keV Result (%)	Destructive Analysis
ASH-1 (standard)	19.79 ± 0.42	19.64 ± 0.44	19.64 ± 0.24	0	19.72
XBLP-267	124.5 ± 0.7	122.5 ± 0.5	118.9 ± 0.3	5	118
XBLP-120	93.7 ± 0.2	92.1 ± 0.6	91.0 ± 0.3	1	105
XBLP-270	95.0 ± 0.5	92.4 ± 0.2	90.6 ± 0.4	3	93
XBLP-300	171.5 ± 2.6	171.2 ± 0.6	166.0 ± 0.2	4	18[illegible]

the traditional corrections for matrix attenuation, branching ratios, and count rate effects have already been made. If the ratio of the results obtained at different energies is 1, there will be no correction. The lump corrections based on the 129/414, 203/414 and 345/414 ratios have been applied to the uncorrected assay results shown in Table I. The corrected results are shown in Table III. We found good agreement between the three independent lump-corrected assay results. The lump correction for these samples ranges from 1 to 5%. Destructive analysis of the samples is also shown in Table III. For the samples XBLP-267 and XBLP-270, the agreement is excellent; for samples XBLP-120 and 300, the lump-corrected SGS assay results are 10% low. Additional destructive analyses on other samples are in progress.

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